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(71) Applicant(s):
Fuji Electric Holdings Company Limited
(Incorporated in Japan)
1-1 Tanabeshinden, Kawasaki-ku,
Kawasaki 210-0858, Japan

(72) Inventor(s):
Hiroshi Kimura

(74) Agent and/or Address for Service:
G F Redfern & Co
Lynn House, Ivy Arch Road, WORTHING,
West Sussex, BN14 8BX, United Kingdom

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Jitsuyo Shinan Koho 1994-2003, Jitsuyo Shinan
Toroku Koho 1996-2003

(54) Abstract Title: Organic el device and organic el panel

(57) An organic EL device wherein a transparent conductive film of one of $\text{In}_2\text{O}_3\text{-ZnO}$, $\text{In}_2\text{O}_3\text{-SnO}_2$, ZnO , and SnO_2 is provided on the light-emitting layer side of a metal electrode of the organic EL device, the thickness of the transparent conductive film is so determined as to satisfy the formula below where L is the optical distance between an organic light-emitting layer and the metal electrode, and λ is the wavelength of the emitted light and the light waves reflected from the metal electrode interfere with one another to strengthen one another. The external quantum efficiency is improved without degrading the luminance, and the contrast can be improved. An organic EL panel using the organic EL device is also disclosed. $L = (2n+1)\lambda/4$ ($n=0, 1, 1, \dots$).

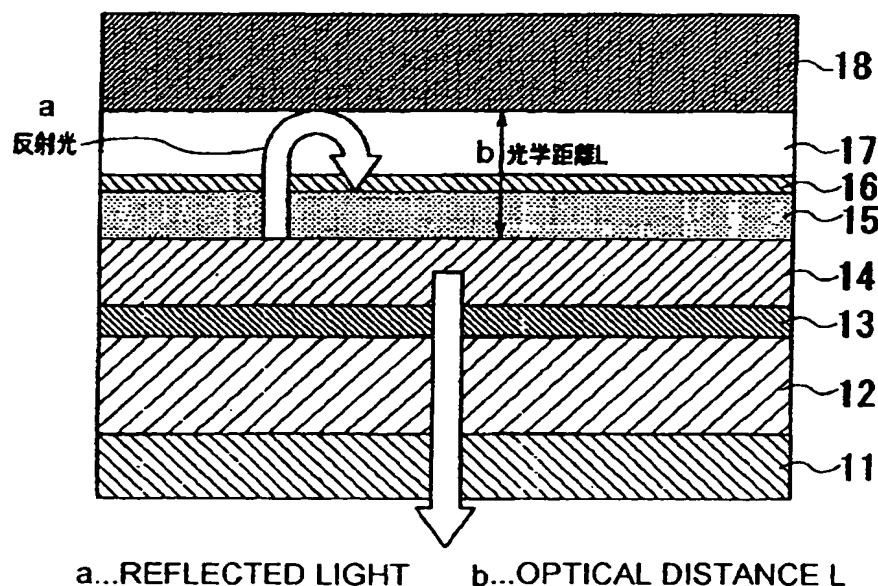


Fig 1

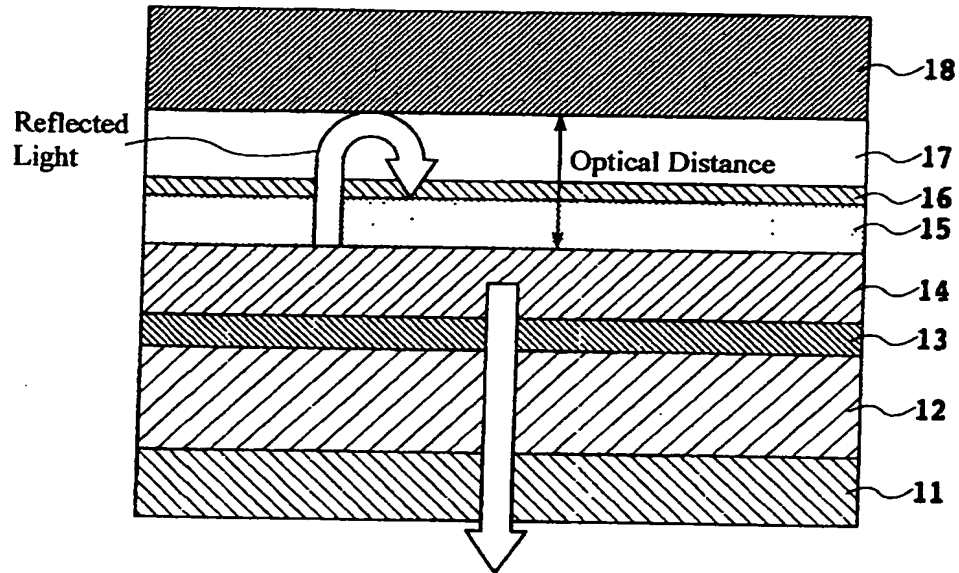


Fig 2

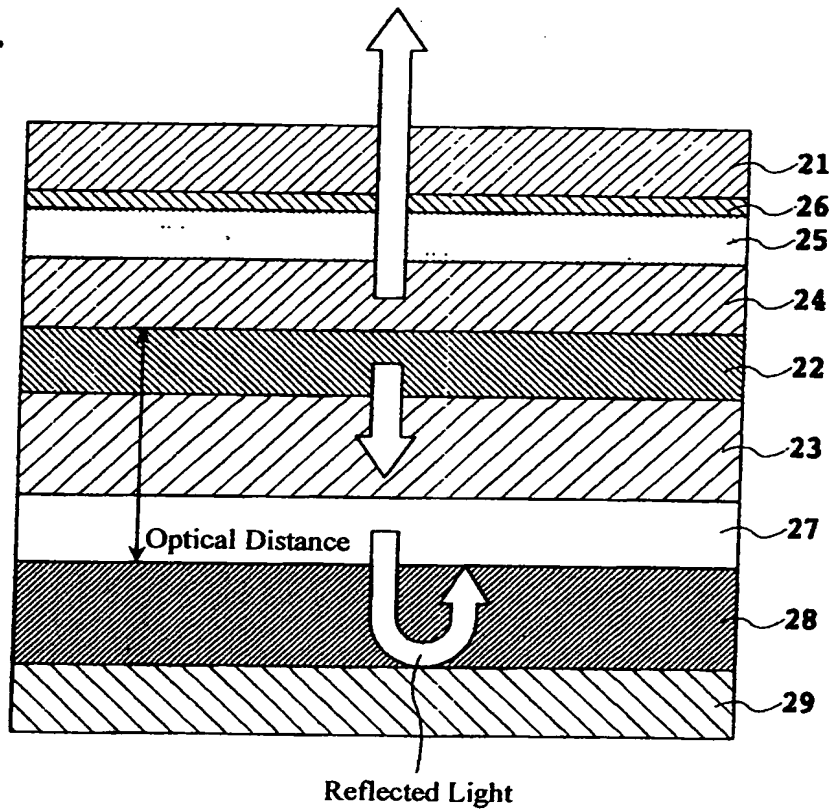


Fig 3

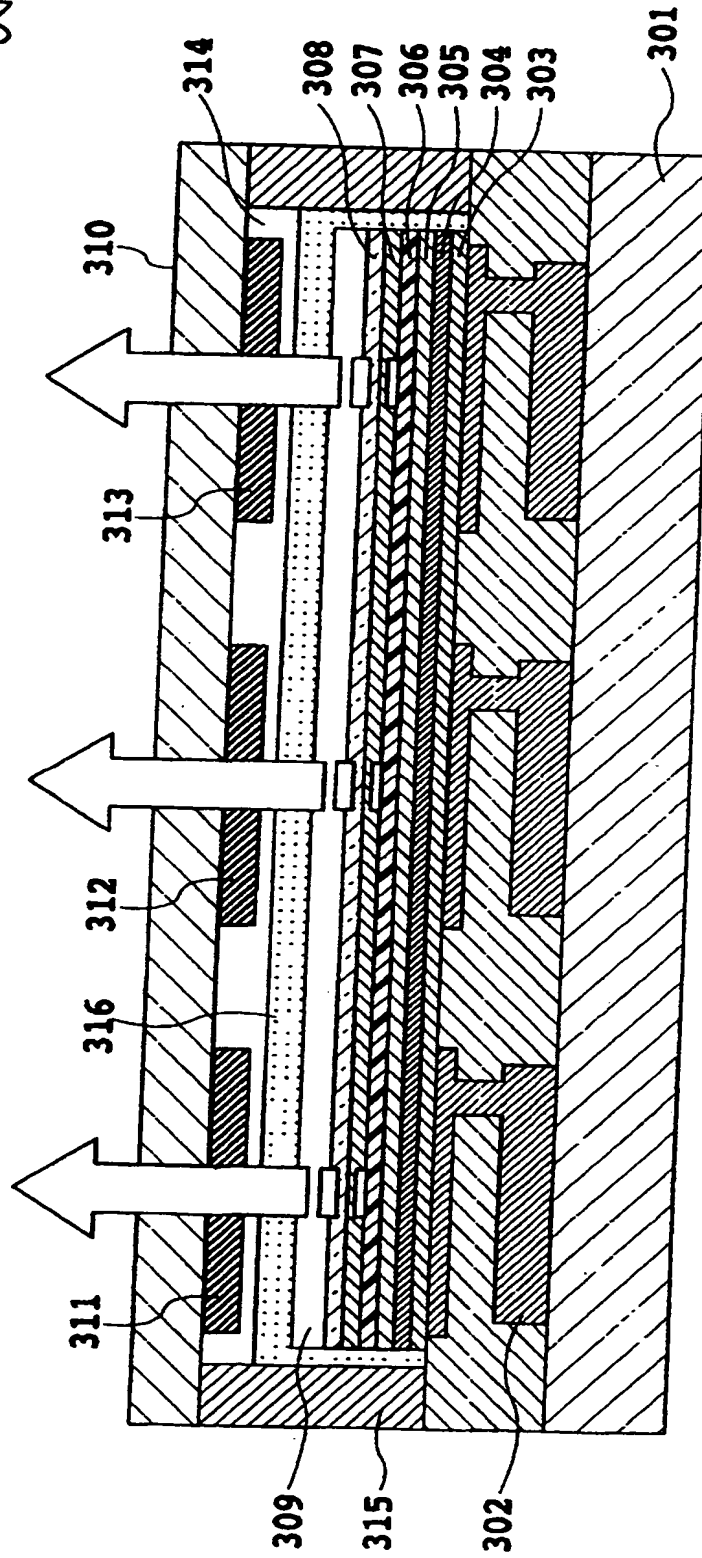
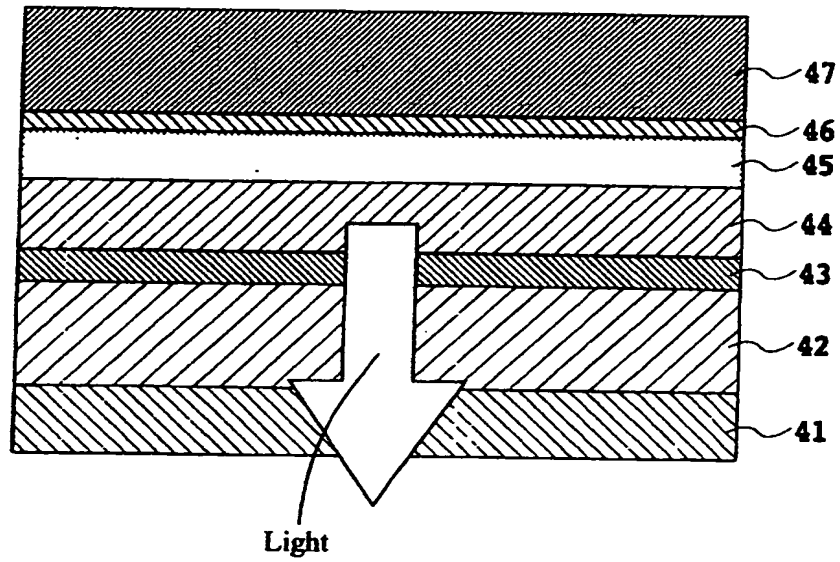


Fig 4



ORGANIC EL DEVICE AND ORGANIC EL PANEL

The present invention relates to an organic EL (electroluminescent) device and an organic EL panel, and more specifically relates to an organic EL device and an organic EL panel using the same, according to which the external quantum efficiency can be improved with no accompanying deterioration in the brightness, and moreover the contrast can be improved.

Ever since the announcement by Tang in 1987 of a high-efficiency organic EL device having a two-layer laminated structure (C.W. Tang et al., Appl. Phys. Lett., 51, 913 (1987)), various organic EL devices have been developed, and some of these have already been put into practical use.

Figure 4 is a drawing for explaining the structure of a conventional organic EL device; the device has a constitution in which a hole injection layer 42, a hole transport layer 43, a light-emitting layer 44, an electron transport layer 45, and an electron injection layer 46 are formed in this order on a transparent electrode 41, which is an anode, and a metal electrode 47, which is a cathode, is provided on the electron injection layer 46.

The quantum efficiency of an organic EL device having a structure as shown in Figure 4 is thought of as follows. First, holes and electrons arriving from the anode and the cathode form electron-hole pairs in the light-emitting layer, thus becoming excitons having light-emitting ability; the

probability of production of such a light-emitting exciton is approximately 25%. The efficiency of extracting light produced in the light-emitting layer out to the outside of the device, on the other hand, is given by the following equation, where n is the refractive index of the light-emitting layer.

$$\chi = \frac{1}{2n^2} \quad (1)$$

Generally, the refractive index of the light-emitting layer is 1.6, and hence the efficiency of extracting out light is approximately 20%. The theoretical external quantum efficiency limit is thus given by the product of the probability of production of a light-emitting exciton (approximately 25%) and the efficiency of extracting out light (approximately 20%), which is approximately 5%.

However, the external quantum efficiency of an actual organic EL device is low at approximately 3%, which is about 60% of the theoretical value, and hence a problem arises in that if the current passed through the device is increased to extract light of a fixed brightness to the outside, then deterioration of the brightness will progress, and in addition the energy consumption will be increased.

Moreover, with actual panels, a problem of contrast in which the display becomes hard to see due to external light is a problem in actual practice. One cause of such a drop in the

contrast is said to be the metal electrode reflecting external light.

In view of such problems, it is an object of the present invention to provide an organic EL device and an organic EL panel using the same, according to which the external quantum efficiency can be improved with no accompanying deterioration in the brightness, and moreover the contrast can be improved.

The present invention has been accomplished to solve the above problems; an invention according to claim 1 is an organic EL device comprising an organic EL light-emitting part including an organic light-emitting layer, between a metal electrode and a transparent electrode, characterised in that a transparent electrically conductive film is provided on a surface of the metal electrode on the organic EL light-emitting part side, and the thickness of the transparent electrically conductive film is set such as to satisfy the following equation, where L is the optical distance from the organic light-emitting layer to the metal electrode, and λ is the wavelength of light emitted by the organic light-emitting layer.

$$L = \frac{2n+1}{4} \lambda \quad (n = 0, 1, 2, \dots) \quad (2)$$

Moreover, an invention according to claim 2 is an organic EL device comprising an organic EL light-emitting part

including an organic light-emitting layer, between a metal electrode and a transparent electrode, characterised in that a transparent electrically conductive film is provided on a surface of the metal electrode on the organic EL light-emitting part side, and light of wavelengths different to the wavelength of light emitted by the organic EL light-emitting layer is absorbed by at least one or both of the metal electrode and the transparent electrically conductive film, and only light of the wavelength emitted by the organic EL light-emitting layer is discharged from the transparent electrode.

Moreover, an invention according to claim 3 is an organic EL device comprising an organic EL light-emitting part including an organic light-emitting layer, between a metal electrode and a transparent electrode, characterised in that a transparent electrically conductive film is provided on a surface of the metal electrode on the organic EL light-emitting part side, the thickness of the transparent electrically conductive film is set such as to satisfy the following equation, where L is the optical distance from the organic light-emitting layer to the metal electrode, and λ is the wavelength of light emitted by the organic light-emitting layer,

$$L = \frac{2n+1}{4} \lambda \quad (n = 0, 1, 2, \dots) \quad (3)$$

and light of wavelengths different to the wavelength of light emitted by the organic EL light-emitting layer is absorbed by the metal electrode and/or the transparent electrically conductive film, and only light of the wavelength emitted by the organic EL light-emitting layer is discharged from the transparent electrode.

Moreover, an invention according to claim 4 is the organic EL device according to any one of claims 1 through 3, characterised in that a material of the transparent electrically conductive film is one of $\text{In}_2\text{O}_3\text{-ZnO}$, $\text{In}_2\text{O}_3\text{-SnO}_2$, ZnO and SnO_2 .

Moreover, an invention according to claim 5 is the organic EL device according to claim 2 or 3, characterised in that the transparent electrically conductive film has an impurity added thereto so as to be coloured to a colour the same as the colour of the light emitted by the organic EL light-emitting layer.

Moreover, an invention according to claim 6 is the organic EL device according to claim 5, characterised in that the organic EL light-emitting layer emits blue light, the transparent electrically conductive film is constituted from a material of one of $\text{In}_2\text{O}_3\text{-ZnO}$, $\text{In}_2\text{O}_3\text{-SnO}_2$, ZnO and SnO_2 ,

containing an impurity of one of CuO, Co and Ti at a concentration of not more than 1%, and the transparent electrically conductive film absorbs blue light.

Moreover, an invention according to claim 7 is the organic EL device according to any one of claims 2, 3 and 6, characterised in that the organic EL light-emitting layer emits blue light, the metal electrode comprises Zn, Mo or Cr, or an alloy thereof, and the metal electrode absorbs blue light.

Moreover, an invention according to claim 8 is a monochrome panel or area colour panel, characterised by comprising the organic EL device according to any one of claims 1 through 5.

Moreover, an invention according to claim 9 is a colour conversion type colour panel, characterised by comprising the organic EL device according to claim 6, a blue monochrome backlight, and colour-converting filters, wherein light other than blue light is absorbed by the transparent electrically conductive film of the organic EL device, and only blue monochrome light from the backlight is reflected by the metal electrode.

Moreover, an invention according to claim 10 is a colour conversion type colour panel, characterised by comprising the organic EL device according to claim 7, a blue monochrome backlight, and colour-converting filters, wherein light other

than blue light is absorbed by the metal electrode, and only blue monochrome light from the backlight is reflected by the metal electrode.

The invention will now be described with reference to the drawings, in which:

Figure 1 is a drawing for explaining an example of the constitution of an organic EL device of the present invention;

Figure 2 is a drawing for explaining a second example of the constitution of an organic EL device of the present invention;

Figure 3 is a sectional view of a colour conversion type colour panel constituted using an organic EL device of the present invention; and

Figure 4 is a drawing for explaining the structure of a conventional organic EL device.

Referring to the Figures, Figure 1 is a drawing for explaining an example of the constitution of an organic EL device of the present invention formed on a substrate; this organic EL device includes an organic EL light-emitting part constituted from a plurality of organic layers including an organic light-emitting layer, and specifically has a constitution in which a hole injection layer 12, a hole transport layer 13, a light-emitting layer 14, an electron transport layer 15, and an electron injection layer 16 are formed in this order on a transparent electrode 11, which is

an anode, a transparent electrically conductive film 17 is provided on the electron injection layer 16, and a metal electrode 18, which is a cathode metal layer, is provided on the transparent electrically conductive film 17. Note that when constituting the organic EL device of the present invention, a glass substrate may be provided on either the transparent electrode 11 (the anode), or the metal electrode 18 (the cathode metal layer).

Of the light emitted from the light-emitting layer 14, the light emitted to the hole transport layer 13 side passes through the hole transport layer 13 and the hole injection layer 12 and is extracted to the outside from the transparent electrode 11, whereas light emitted to the electron transport layer 15 side passes through the electron transport layer 15, the electron injection layer 16 and the transparent electrically conductive film 17, and is reflected by the metal electrode 18 and thus returns into the device. Consequently, if this reflected light can be extracted to the outside without being weakened in the device, then the external quantum efficiency can be improved.

That is, taking the thicknesses and refractive indices of the electron transport layer 15, the electron injection layer 16 and the transparent electrically conductive film 17 in the device to be d_i ($i = 1, 2, 3$) and n_i ($i = 1, 2, 3$) respectively, the optical distance L from the light-emitting

layer 14 to the metal electrode 18 will be given by the sum of the optical distances for these layers, i.e. by the following equation.

$$L = \sum_i n_i d_i \quad (4)$$

When light is reflected at the interface between the metal electrode 18 and the transparent electrically conductive film 17, the phase of the light is reversed, and hence taking the wavelength of the light to be λ , a condition for the light strengthening itself in the device is:

$$L = \frac{2n+1}{4} \lambda \quad (n = 0, 1, 2, \dots) \quad (5)$$

Because the metal electrode 18 is used as the cathode, and the electron transport layer 15, the electron injection layer 16 and the transparent electrically conductive film 17 are interposed between the metal electrode 18 and the light-emitting layer 14, if design is carried out such that the optical distance accounted for by these layers satisfies equation (5), then the external quantum efficiency can be improved.

However, the thickness of the electron injection layer 16 must be made low at approximately 0.5 to 1 nm, and moreover

if the thickness of the electron transport layer 15 is made to be high then there will be a problem of deterioration of the brightness of the device becoming marked; consequently, with the organic EL device of the present invention, the transparent electrically conductive film 17 is provided between the electron injection layer 16 and the metal electrode 18, and the thickness of the transparent electrically conductive film 17 is set such that the light reflected by the metal electrode 18 satisfies the above interference condition, whereby the light can be extracted to the outside without the strength of the light being weakened in the device, and hence the external quantum efficiency can be improved.

The method of setting the optical distance such that the external quantum efficiency is maximized by adjusting the thickness of the transparent electrically conductive film 17 in this way is not only effective with a monochrome panel or area colour panel that is made to emit light using a monochrome backlight, but moreover is particularly effective with a colour panel that uses a colour conversion method in which light emitted from a monochrome backlight is received by colour-converting layers and converted into red, green and blue light.

Moreover, one practical problem of organic EL panels is a drop in contrast due to external light, and it has been

ascertained that a cause of this is external light being directly reflected by the metal electrode. According to equation (5), there is limited light of wavelengths strengthened by interference, with only light of specific wavelengths being reflected; the strength of reflection of light of wavelengths not satisfying equation (5) will thus be reduced, and hence it can be seen that the organic EL device of the present invention contributes to improving the contrast of an organic EL panel.

To further improve the contrast, it is effective to form the transparent electrically conductive film and the metal layer on top of one another to constitute a reflective layer, and colour the transparent electrically conductive film out of this reflective layer to the colour of the emitted light, thus producing a structure according to which light of colours other than the colour of the emitted light cannot be reflected, or make the material of the metal layer be a material having a property of absorbing light of colours other than the colour of the emitted light. One can thus envisage a method in which light of wavelengths not required to be extracted out from the transparent electrode is absorbed by the laminate of the transparent electrically conductive film and the metal layer, or a method in which this light is absorbed by the metal layer material. Note that, in this case, it is preferable to constitute the various layers such

that the optical distance for the layers interposed between the metal electrode and the light-emitting layer satisfies equation (5), but there is no limitation to this.

In particular, with a colour conversion type colour panel, the backlight is blue, and hence it is effective to use a metal having a larger reflection coefficient for blue than for red as the reflective metal; specifically, Zn, Mo or Cr may be used. Moreover, the transparent electrically conductive film can be made blue by, for example, adding CuO, Co or Ti in an amount of not more than 1% to the oxide layer constituting the transparent electrically conductive film.

In addition to the constitution shown in Figure 1, an organic EL device of the present invention may have the constitution shown in Figure 2.

Figure 2 is a drawing for explaining the structure in the case that the lower electrode of the organic EL device is made to be an anode; with this constitution, a metal electrode 28, a transparent electrically conductive film 27, which is an anode, a hole injection layer 23, a hole transport layer 22, a light-emitting layer 24, an electron transport layer 25, an electron injection layer 26, and a transparent electrode 21, which is a cathode, are formed in this order on a substrate 29. Here, regarding the constitution of the part comprising the electron injection layer 26 and the transparent electrode 21 (the cathode), one can envisage a structure in which the

electron injection layer 26 is formed from an ultra-thin film of an alkali or alkaline earth metal oxide, fluoride, boride or chloride, an ultra-thin film of a metal such as Al is deposited thereon, and an $\text{In}_2\text{O}_3\text{-ZnO}$ oxide layer (IZO) is further provided thereon, or a structure in which a transparent electrode 21 made of a transparent oxide such as IZO is directly deposited on the electron injection layer 26.

Note that in addition to organic EL devices having the layer structures shown in Figures 1 and 2, the present invention can also be applied to organic EL devices having all other organic EL device constitutions proposed hitherto, for example a constitution in which a hole transport layer is not provided.

Example 1

Figure 3 is a sectional view of a colour conversion type colour panel constituted using an organic EL device of the present invention. Cr (5 nm) and Pt (100 nm) were deposited as a metal electrode 303 as reflective metal on a substrate 301 having TFTs 302 thereon, and then an $\text{In}_2\text{O}_3\text{-ZnO}$ oxide layer (IZO: refractive index 2.2) was further deposited as a transparent electrically conductive film 304 acting as an anode on the metal electrode 303. The metal electrode 303 used here as the reflective metal is not limited to being a laminate of Cr and Pt so long as it is an electrically conductive metal or alloy having unevenness of not more than 4

nm. Moreover, the formation of the IZO film was carried out by sputtering, but may instead be carried out using another film formation method such as electron beam vapour deposition or resistive heating vapour deposition.

A hole injection layer 305, a hole transport layer 306, and a light-emitting layer 307 were deposited in this order on the transparent electrically conductive film 304 by resistive heating vapour deposition, and then an 8-hydroxyquinoline Al complex (Alq_3) was further formed thereon to a thickness of 20 nm as an electron transport layer 308.

A laminate 309 of an electron injection layer and an upper transparent electrode was constituted by depositing LiF to a thickness of 0.5 nm as the electron injection layer, then depositing 1 nm of Al and 220 nm of IZO as the upper transparent electrode, and then finally depositing 300 nm of SiON as a protective layer.

The optical distance for the organic EL device having this constitution was adjusted between the IZO transparent electrically conductive film 304 that was the lower electrode (the anode), the hole injection layer 305, the hole transport layer 306, and the Pt film in the metal electrode 303. The wavelength of the light from the colour conversion type backlight was 470 nm, and the hole injection layer 305 was deposited to a thickness of 80 nm and the hole transport layer 306 to a thickness of 20 nm; consequently, taking the

refractive index of the organic material to be 1.85, from the interference condition of equation (5) the IZO film thickness was made to be 183 nm. Furthermore, the IZO film that was the transparent electrically conductive film 304 constituting the lower electrode was coloured blue by adding 0.6% of CuO thereto.

A protective layer 316 was provided on the substrate 301 on which the device had been formed as described above, and this substrate and a substrate 310 on which red, green and blue colour-converting filters 311, 312 and 313 had been formed in advance were put face to face, and then with a gap therebetween filled with a gel body 314, an outer peripheral portion of the device was sealed with an outer periphery sealant 315, thus completing the panel. Here, 'colour-converting filters' are filters provided with fluorescent filters and/or colour filters.

As a result of comparing the properties of the panel having the constitution described in the present example with the properties of a panel having a conventional constitution, it was found that the efficiency of extracting out light could be improved from 2.0% to 3.0%, and hence the current passed at the same brightness could be reduced to 2/3. Furthermore, a contrast ratio of 200:1 at 100 cd/m² under 1000 Lx was obtained.

Moreover, upon carrying out similar comparative experiments with monochrome and area colour panels, similar results were obtained.

Example 2

The same comparison as in Example 1 was carried out but using $\text{In}_2\text{O}_3\text{-SnO}_2$ (ITO) (refractive index 2.0) of film thickness 201 nm instead of $\text{In}_2\text{O}_3\text{-ZnO}$ as the transparent electrically conductive film material; in this case, similar effects to with Example 1 were again obtained. The ITO film can be deposited using a method such as sputtering, vapour deposition or CVD. Moreover, similar results were also obtained in the case of using ZnO or SnO_2 as the transparent electrically conductive film material and adjusting the optical distance.

As described above, according to the present invention, a transparent electrically conductive film is provided on a surface of a metal electrode of an organic EL device on the light-emitting layer side, and the thickness of this transparent electrically conductive film is adjusted such that light reflected by the metal electrode is made to undergo interference and thus strengthen itself in the device, whereby the external quantum efficiency can be improved with no accompanying deterioration in the brightness; furthermore, light of a specific wavelength is made to be absorbed by the metal electrode and the transparent electrically conductive film, and hence the contrast is improved; as a result, there

can be provided an organic EL device and an organic EL panel using the same, according to which the external quantum efficiency can be improved with no accompanying deterioration in the brightness, and moreover the contrast can be improved.

CLAIMS

1. An organic EL device comprising an organic EL light-emitting part including an organic light-emitting layer, between a metal electrode and a transparent electrode, the organic EL device characterised in that:

a transparent electrically conductive film is provided on a surface of the metal electrode on the organic EL light-emitting part side;

and the thickness of the transparent electrically conductive film is set such as to satisfy the following equation, where L is the optical distance from the organic light-emitting layer to the metal electrode, and λ is the wavelength of light emitted by the organic light-emitting layer.

$$L = \frac{2n+1}{4} \lambda \quad (n = 0, 1, 2, \dots) \quad (6)$$

2. An organic EL device comprising an organic EL light-emitting part including an organic light-emitting layer, between a metal electrode and a transparent electrode, the organic EL device characterised in that:

a transparent electrically conductive film is provided on a surface of the metal electrode on the organic EL light-emitting part side;

and light of wavelengths different to the wavelength of light emitted by the organic light-emitting layer is absorbed by at least one or both of the metal electrode and the transparent electrically conductive film, and only light of the wavelength emitted by the organic EL light-emitting layer is discharged from the transparent electrode.

3. An organic EL device comprising an organic EL light-emitting part including an organic light-emitting layer, between a metal electrode and a transparent electrode, the organic EL device characterised in that:

a transparent electrically conductive film is provided on a surface of the metal electrode on the organic EL light-emitting part side;

the thickness of the transparent electrically conductive film is set such as to satisfy the following equation, where L is the optical distance from the organic light-emitting layer to the metal electrode, and λ is the wavelength of light emitted by the organic light-emitting layer,

$$L = \frac{2n+1}{4} \lambda \quad (n = 0, 1, 2, \dots) \quad (7)$$

and light of wavelengths different to the wavelength of light emitted by the organic EL light-emitting layer is absorbed by the metal electrode and/or the transparent electrically conductive film, and only light of the wavelength emitted by the organic EL light-emitting layer is discharged from the transparent electrode.

4. The organic EL device according to any one of claims 1 through 3, characterised in that a material of the transparent electrically conductive film is one of $\text{In}_2\text{O}_3\text{-ZnO}$, $\text{In}_2\text{O}_3\text{-SnO}_2$, ZnO and SnO_2 .
5. The organic EL device according to claim 2 or 3, characterised in that the transparent electrically conductive film has an impurity added thereto so as to be coloured to a colour the same as the colour of the light emitted by the organic EL light-emitting layer
6. The organic EL device according to claim 5, characterised in that the organic EL light-emitting layer emits blue light, the transparent electrically conductive film is constituted from a material of one of $\text{In}_2\text{O}_3\text{-ZnO}$, $\text{In}_2\text{O}_3\text{-SnO}_2$, ZnO and SnO_2 , containing an impurity of one of CuO , Co and Ti at a concentration of not more than 1%, and the transparent electrically conductive film absorbs blue light.

7. The organic EL device according to any one of claims 2, 3 and 6, characterised in that the organic EL light-emitting layer emits blue light, the metal electrode comprises Zn, Mo or Cr, or an alloy thereof, and the metal electrode absorbs blue light.
8. A monochrome panel or area colour panel, characterised by including the organic EL device according to any one of claims 1 through 5.
9. A colour conversion type colour panel, characterised by comprising the organic EL device according to claim 6, a blue monochrome backlight, and colour-converting filters, wherein light other than blue light is absorbed by the transparent electrically conductive film of the organic EL device, and only blue monochrome light from the backlight is reflected by the metal electrode.
10. A colour conversion type colour panel, characterised by comprising the organic EL device according to claim 7, a blue monochrome backlight, and colour-converting filters, wherein light other than blue light is absorbed by the metal electrode, and only blue monochrome light from the backlight is reflected by the metal electrode.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP03/07565

A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl⁷ H05B33/28

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl⁷ H05B33/28

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho	1922-1996	Toroku Jitsuyo Shinan Koho	1994-2003
Kokai Jitsuyo Shinan Koho	1971-2003	Jitsuyo Shinan Toroku Koho	1996-2003

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	WO 99/39393 A (INT BUSINESS MACHINES CORP.), 05 August, 1999 (05.08.99), Full text & JP 2001-507167 A	1-10
Y	JP 2002-289358 A (Ricoh Co., Ltd.), 04 October, 2002 (04.10.02), Full text (Family: none)	1-10

☐ Further documents are listed in the continuation of Box C.☐ See patent family annex.

* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A" document defining the general state of the art which is not considered to be of particular relevance	"X" document of particular relevance, the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E" earlier document but published on or after the international filing date	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
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"O" document referring to an oral disclosure, use, exhibition or other means	
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Date of the actual completion of the international search
22 September, 2003 (22.09.03)Date of mailing of the international search report
07 October, 2003 (07.10.03)Name and mailing address of the ISA/
Japanese Patent Office

Authorized officer

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Telephone No.